# Quantum Field Nature of Relaxation by Strong Coupling in Joint Electromagnetic Field - Dipole Moments - Phonon Field System

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QED-model for dynamics of spectroscopic transitions in 1D multiqubit exchange coupled system, presented in [14] is generalized by taking into account more carefully quantum nature of relaxation processes. The formation of coherent system of the resonance phonons is predicted and experimentally confirmed. It leads to oscillation character of quantum relaxation, that was experimentally identified for the first time.

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### I. INTRODUCTION AND BACKGROUND

Quantum electrodynamics (QED) takes on more and more significance for its practical application and it, in fact, becomes to be working instrument in spectroscopy studies and industrial spectroscopy control. Quantum dynamics of two level systems (qubits), coupled to a single mode of an electromagnetic cavity are of considerable interest in connection with quickly developing new quantum physics branches like to cavity quantum electrodynamics [1] or quantum computing [2]. Spin-based solid state quantum bits are known to have long coherence times, while also offering the promise of scalability, and are natural building blocks for quantum computation. Phosphorus donor nuclei in silicon have been known since the 1950s to have some of the best spin coherence properties in solids. The spin coherence time  $T_2$ measured by Hahn spin echo method for donor electron spins in bulk Si:P has been reported to be equal  $\sim 60ms$ [3]. This is the longest coherence time measured in electron spin qubits, and greatly exceeds the value reported, for instance, in GaAs quantum dots, which, measured also by Hahn spin echo method, is equal to  $\sim 100 \mu s$  [4]. However, fabrication of ordered and gated donor arrays and coherent control over donor electrons has turned out to be extremely difficult. There is now much interest in fabricating spin-based devices in diamond, with potential applications in quantum communication, quantum computation, and magnetometry. Nitrogen-vacancy (NV) centers located deep within a diamond lattice appear promising solid-state spin qubits since they combine optical initialization and readout capabilities owing to long electron spin coherence life times approaching 1ms at room temperature [5], and the ability to control coupling to individual nuclear spins. In all of aforesaid applications it is necessary or at least advantageous to couple NV centers to optical structures like to waveguides and microresonators, to enable communication between distant qubits or to allow efficient extraction of emitted photons. Therefore, a reliable method is needed to create NV centers with good spectral properties in close proximity ( $\lesssim 100nm$ ) to a diamond surface. In addition, the charge state of the NV center must be controlled. Given tasks are under study at present. We will show in present work another way to obtain long-lived coherent states with similar field of practical application. The prediction will be based on QED-theory of spectroscopic transitions. The simplest models which capture the salient features of the relevant physics in given field are the Jaynes-Cummings model (JCM) [6] for the one qubit case and its generalization for multiqubit systems by Tavis and Cummings [7]. Tavis-Cummings model was generalized in [9], by taking into account the 1D-coupling between qubits. Recently QED-model for one chain coupled qubit system was generalized for quasionedimensional axially symmetric multichain coupled qubit system [8]. It is substantial, that in the model, proposed in [8] the interaction of quantized EM-field with multichain qubit system is considered by taking into account both the intrachain and interchain qubit coupling without restriction on their values. It follows from theoretical results in [9], [8] and from their experimental confirmation in [13], that by strong interaction of EM-field with matter the correct description of spectroscopic transitions including stationary spectroscopy is achieved the only in the frame of QED consideration. It concerns both optical and radio spectroscopies, that means, that QED consideration has to be also undertaken by electron spin resonance (ESR) studies in the case of strong interaction of EM-field with spin systems. It is reasonable to suggest, that analogous conclusion can be drawn for the case of strong interaction of phonon field with spin system or electron system. In other words it seems to be reasonable the idea, that relaxation of paramagnetic (or optical) centers in the case of strong spin-phonon (electron-phonon) interaction can be described correctly the only in the frames of quantum field theory.

The aim of the work presented is to show theoretically and experimentally, that in the case aforesaid the relaxation process of excited matter electronic or matter spin subsystem by resonance interaction with EM-field, in particular by ESR studies, can really display its quan-

tum nature and to play the essential role in dynamics of spectroscopic transitions, including radiospectroscopy.

#### II. RESULTS AND DISCUSSION

## A. QED Equations for Spectroscopic Transitions by Strong Electron-Phonon Coupling

Recently in the work [14] the system of QED differencedifferencial equations for dynamics of spectroscopic transitions for both radio- and optical spectroscopy for the model, representing itself the 1D-chain of N two-level equivalent elements interacting with quantized EM-field (that is N qubits), coupled by exchange interaction (or its optical analogue for the optical transitions) have recently been derived. Naturally the equations are true for any 3D system of paramagnetic centers (PC) or optical centers by the absence of exchange interaction. In given case the model presented differs from Tavis-Cummings model [7] by inclusion into consideration of quantized phonon system, describing the relaxation processes from quantum fied theory position. Seven equations for the seven operator variables, describing joint system {field + matter} can be presented in matrix form by three matrix equations. They are the following

$$\frac{\partial}{\partial t} \begin{bmatrix} \hat{\sigma}_l^- \\ \hat{\sigma}_l^+ \\ \hat{\sigma}_l^z \end{bmatrix} = 2 \|g\| \begin{bmatrix} \hat{F}_l^- \\ \hat{F}_l^+ \\ \hat{F}_l^z \end{bmatrix} + \|\hat{R}_{\vec{q}l}^{(\lambda)}\|, \tag{1}$$

$$\frac{\partial}{\partial t} \begin{bmatrix} \hat{a}_{\vec{k}} \\ \hat{a}_{\vec{k}}^{+} \end{bmatrix} = -i\omega_{\vec{k}} ||\sigma_{P}^{z}|| \begin{bmatrix} \hat{a}_{\vec{k}} \\ \hat{a}_{\vec{k}}^{+} \end{bmatrix}$$

$$+\frac{i}{\hbar} \begin{bmatrix} -\sum_{l=1}^{N} (\hat{\sigma}_{l}^{+} + \hat{\sigma}_{l}^{-}) v_{l\vec{k}}^{*} \\ \sum_{l=1}^{N} (\hat{\sigma}_{l}^{+} + \hat{\sigma}_{l}^{-}) v_{l\vec{k}} \end{bmatrix},$$
 (2)

$$\frac{\partial}{\partial t} \begin{bmatrix} \hat{b}_{\vec{k}} \\ \hat{b}_{\vec{q}}^{+} \end{bmatrix} = -i\omega_{\vec{q}} ||\sigma_{P}^{z}|| \begin{bmatrix} \hat{b}_{\vec{q}} \\ \hat{b}_{\vec{q}}^{+} \end{bmatrix} + \frac{i}{\hbar} \begin{bmatrix} -\sum_{l=1}^{N} \hat{\sigma}_{l}^{z} \lambda_{\vec{q}l} \\ \sum_{l=1}^{N} \hat{\sigma}_{l}^{z} \lambda_{\vec{q}l} \end{bmatrix} (3)$$

where

$$\begin{bmatrix} \hat{\sigma}_l^- \\ \hat{\sigma}_l^+ \\ \hat{\sigma}_l^z \end{bmatrix} = \hat{\vec{\sigma}}_l = \hat{\sigma}_l^- \vec{e}_+ + \hat{\sigma}_l^+ \vec{e}_- + \hat{\sigma}_l^z \vec{e}_z$$
 (4)

is vector-operator of spectroscopic transitions for lth chain unit,  $l=\overline{2,N-1}$  [14]. Its components, that is, the operators

$$\hat{\sigma}_v^{jm} \equiv |j_v\rangle\langle m_v| \tag{5}$$

are set up in correspondence to the states  $|j_v\rangle,\langle m_v|$ , where  $v=\overline{1,N},\ j=\alpha,\beta,\ m=\alpha,\beta$ . For instance, the relationships for commutation rules are

$$[\hat{\sigma}_v^{lm}, \hat{\sigma}_v^{pq}] = \hat{\sigma}_v^{lq} \delta_{mp} - \hat{\sigma}_v^{pm} \delta_{ql}. \tag{6}$$

Further

$$\begin{bmatrix} \hat{F}_{l}^{-} \\ \hat{F}_{l}^{+} \\ \hat{F}_{l}^{z} \end{bmatrix} = \hat{\vec{F}} = \left[ \hat{\vec{\sigma}}_{l} \otimes \hat{\vec{\mathcal{G}}}_{l-1,l+1} \right], \tag{7}$$

where vector operators  $\hat{\vec{\mathcal{G}}}_{l-1,l+1}$ ,  $l = \overline{2, N-1}$ , are given by the expressions

$$\hat{\vec{\mathcal{G}}}_{l-1,l+1} = \hat{\mathcal{G}}_{l-1,l+1}^{-}\vec{e}_{+} + \hat{\mathcal{G}}_{l-1,l+1}^{+}\vec{e}_{-} + \hat{\mathcal{G}}_{l-1,l+1}^{z}\vec{e}_{z}, \quad (8)$$

in which

$$\hat{\mathcal{G}}_{l-1,l+1}^{-} = -\frac{1}{\hbar} \sum_{\vec{k}} \hat{f}_{l\vec{k}} - \frac{J}{\hbar} (\hat{\sigma}_{l+1}^{-} + \hat{\sigma}_{l-1}^{-}), \tag{9a}$$

$$\hat{\mathcal{G}}_{l-1,l+1}^{+} = -\frac{1}{\hbar} \sum_{\vec{k}} \hat{f}_{l\vec{k}} - \frac{J}{\hbar} (\hat{\sigma}_{l+1}^{+} + \hat{\sigma}_{l-1}^{+}), \tag{9b}$$

$$\hat{\mathcal{G}}_{l-1,l+1}^{z} = -\omega_{l} - \frac{J}{\hbar} (\hat{\sigma}_{l+1}^{z} + \hat{\sigma}_{l-1}^{z}). \tag{9c}$$

Here operator  $\hat{f}_{l\vec{k}}$  is

$$\hat{f}_{l\vec{k}} = v_{l\vec{k}} \hat{a}_{\vec{k}} + \hat{a}_{\vec{k}}^{+} v^{*}_{l\vec{k}}. \tag{10}$$

In relations (9) J is the exchange interaction constant in the case of magnetic resonance transitions or its optical analogue in the case of optical transitions, the function  $v_{l\vec{k}}$  in (10) is

$$v_{l\vec{k}} = -\frac{1}{\hbar} p_l^{jm} (\vec{e}_{\vec{k}} \cdot \vec{e}_{\vec{P}_l}) \mathfrak{E}_{\vec{k}} e^{-i\omega_{\vec{k}}t + i\vec{k}\vec{r}}, \tag{11}$$

where  $p_l^{jm}$  is matrix element of operator of magnetic (electric) dipole moment  $\vec{P}_l$  of l-th chain unit between the states  $|j_l\rangle$  and  $|m_l\rangle$  with  $j\in\{\alpha,\beta\}$ ,  $m\in\{\alpha,\beta\}$ ,  $j\neq m$ ,  $\vec{e}_{\vec{k}}$  is unit polarization vector,  $\vec{e}_{\vec{P}_l}$  is unit vector along  $\vec{P}_l$ -direction,  $\mathfrak{E}_{\vec{k}}$  is the quantity, which has the dimension of magnetic (electric) field strength,  $\vec{k}$  is quantized EM-field wave vector, the components of which get a discrete set of values,  $\omega_{\vec{k}}$  is the frequency, corresponding to  $\vec{k}$ th mode of EM-field,  $\hat{a}_{\vec{k}}^+$  and  $\hat{a}_{\vec{k}}$  are EM-field creation and annihilation operators correspondingly. In the suggestion, that the contribution of spontaneous emission is

relatively small, we will have  $p_l^{jm} = p_l^{mj} \equiv p_l$ , where  $j \in \{\alpha, \beta\}, m \in \{\alpha, \beta\}, j \neq m$ . Further matrix  $||\hat{R}_{\vec{a}l}^{(\lambda)}||$  is

$$||\hat{R}_{\vec{q}l}^{(\lambda)}|| = \frac{1}{i\hbar} \begin{bmatrix} 2\hat{B}_{\vec{q}l}^{(\lambda)}\hat{\sigma}_l^- \\ -2\hat{B}_{\vec{q}l}^{(\lambda)}\hat{\sigma}_l^+ \\ 0 \end{bmatrix}$$
(12)

Here  $\hat{B}_{\vec{q}l}^{(\lambda)}$  is

$$\hat{B}_{\vec{q}l}^{(\lambda)} = \sum_{\vec{q}} \lambda_{\vec{q}l} (\hat{b}_{\vec{q}}^{+} + \hat{b}_{\vec{q}}), \tag{13}$$

 $\hat{b}_{\vec{q}}^{+}$  ( $\hat{b}_{\vec{q}}$ ) is the creation (annihilation) operator of the phonon with impulse  $\vec{q}$  and with energy  $\hbar\omega_{\vec{q}}$ ,  $\lambda_{\vec{q}l}$  is electron-phonon coupling constant. In equations (2) and (3)  $\|\sigma_{p}^{z}\|$  is Pauli z-matrix,  $\|g\|$  in equation (1) is diagonal matrix, numerical values of its elements are dependent on the basis choice. It is at appropriate basis

$$||g|| = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}. \tag{14}$$

Right hand side expression in (7) is vector product of vector operators. It can be calculated by using of known expression (15) with additional coefficient  $\frac{1}{2}$  the only, which is appeared, since

$$\left[\hat{\vec{\sigma}}_{l} \otimes \hat{\vec{\mathcal{G}}}_{l-1,l+1}^{-1}\right] = \frac{1}{2} \begin{vmatrix} \vec{e}_{-} \times \vec{e}_{z} & \hat{\sigma}_{l}^{-} & \hat{\mathcal{G}}_{l-1,l+1}^{-} \\ \vec{e}_{z} \times \vec{e}_{+} & \hat{\sigma}_{l}^{+} & \hat{\mathcal{G}}_{l-1,l+1}^{+} \\ \vec{e}_{+} \times \vec{e}_{-} & \hat{\sigma}_{l}^{z} & \hat{\mathcal{G}}_{l-1,l+1}^{z} \end{vmatrix}', \quad (15)$$

the products of two components of two vector operators are replaced by anticommutators of corresponding components. Given detail is mapped by symbol  $\otimes$  in (7) and by symbol ' in determinant (15).

It follows from comparison with semiclassical Landau-Lifshitz (L-L) equation for dynamics of spectroscopic transitions in a chain of exchange coupled centers [15], [14], that the equation, which is given by (1) is its QED-generalization. In comparison with semiclassical description, where the description of dynamics of spectroscopic transitions is exhausted by one vector equation (L-L equation or L-L based equation), in the case of completely quantum consideration L-L type equation describes the only one subsystem of three-part-system, which consist of EM-field, dipole moments' (magnetic or electric) matter subsystem and phonon subsystem. It was concluded in [14], that the presence of additional equations for description of transition dynamics by QED model in comparison with semiclassical model leads to a number of new effects, which can be predicted the only by QED consideration of resonance transition phenomena. One of new effect was described in [14], starting the only from the mathematical structure of the equations. It was argued, that the equations (1), (2) represent

themselves vector-operator difference-differential generalization of the system, which belongs to well known family of equation systems - Volterra model systems, widely used in biological tasks of population dynamics studies, which in its turn is generalization of Verhulst equation. In other words, it was predicted, for instance, that by some parameters in two-sybsystem Volterra model the stochastic component in solution will be appeared. Given prediction has experimental confirmation by the study of optical properties in carbynes [16] indicating, that in given material strong electron-photon interaction is realized, which allows to explain the posibility to observe the stationary IR-reflection or absorbtion spectra both in usual and in stochastic regime.

The terms like to right hand side terms in (3) were used in so called "spin-boson" Hamiltonian [17] and in so called "independent boson model" [18]. Given models were used to study phonon effects in a single quantum dot within a microcavity [19], [20], [21], [22], [23]. So, it has been shown in [22], [23], that the presence of the term in Hamiltonian [14]

$$\hat{\mathcal{H}}^{CPh} = \sum_{j=1}^{N} \sum_{\vec{q}} \lambda_{\vec{q}} (\hat{b}_{\vec{q}}^{+} + \hat{b}_{\vec{q}}) \hat{\sigma}_{j}^{z}, \tag{16}$$

which coincides with corresponding term in Hamiltonian in [22], [23] at N = 1 [contribution of given term to the equations for spectroscopic transitions is  $\pm \sum_{l=1}^{N} \hat{\sigma}_{l}^{z} \lambda_{\vec{q}}$ , see equation (3), (note that the equations for spectroscopic transitions were not derived in above cited works [19], [20], [21], [22], [23])] leads the only to exponential decrease of the magnitude of quantum Rabi oscillations with increase of electron-phonon coupling strength and even to their supression at relatively strong electron-phonon coupling. However by strong electronphoton coupling amd strong electron-phonon coupling quite other picture of quantum relaxation processes becomes to be possible. Really, if to define the wave function of the chain system, interacting with quantized EMfield and with phonon field, to be vector of the state in Hilbert space over quaternion ring, that is quaternion function of quaternion argument, then like to [14] can be shown, that the equations (1) to (3) are Lorentz invariant and the transfer to observables can be realized. In particular, taking into account, that quaternion vector of the state is proportional to spin, the Hamiltonian, given by (16) describes in fact the interaction of phonon field with z- component  $S^z$  of the spin of matter subsystem. It seems to be reasonable to take into consideration the interaction of phonon field with  $S^+$ - and  $S^-$  components of the spin of matter subsystem. Therefore we come in a natural way to the following Hamiltonian

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}^C + \hat{\mathcal{H}}^F + \hat{\mathcal{H}}^{CF} + \hat{\mathcal{H}}^{Ph} + \hat{\mathcal{H}}^{CPh}, \tag{17}$$

where  $\hat{\mathcal{H}}^C$  is chain Hamiltonian by the absence of the interaction with EM-field,  $\hat{\mathcal{H}}^F$  is field Hamiltonian,  $\hat{\mathcal{H}}^{CF}$ 

is Hamiltonian, describing the interaction between quantized EM-field and atomic chain. Hamiltonian  $\hat{\mathcal{H}}^C$  is

$$\hat{\mathcal{H}}^C = \hat{\mathcal{H}}^0 + \hat{\mathcal{H}}^J,\tag{18}$$

where  $\hat{\mathcal{H}}^0$  is chain Hamiltonian in the absence of the interaction between structural elementary units of the chain.  $\hat{\mathcal{H}}^0$  is given by the expression

$$\hat{\mathcal{H}}^0 = \sum_{v=1}^N \sum_m E_{mv} |m_v\rangle\langle m_v|. \tag{19}$$

Here  $m = \alpha, \beta$ ,  $E_{mv}$  are eigenvalues of  $\hat{\mathcal{H}}^0$ , which correspond to the states  $|m_v\rangle$  of vth chain unit. Hamiltonian  $\hat{\mathcal{H}}^J$  is

$$\hat{\mathcal{H}}^{J} = \sum_{n=1}^{N} [J_{E}(\hat{\sigma}_{n}^{+}\hat{\sigma}_{n+1}^{-} + \hat{\sigma}_{n}^{-}\hat{\sigma}_{n+1}^{+} + \frac{1}{2}\hat{\sigma}_{n}^{z}\hat{\sigma}_{n+1}^{z}) + H.c.].$$

It is suggested in the model, that  $|\alpha_n\rangle$  and  $|\beta_n\rangle$  are eigenstates, producing the full set for each of N elements. It is evident, that given assumption can be realized strictly the only by the absence of the interaction between the elements. At the same time proposed model will rather well describe the real case, if the interaction energy of adjacent elements is much less of the energy of the splitting  $\hbar\omega_0 = \mathcal{E}_\beta - \mathcal{E}_\alpha$  between the energy levels, corresponding to the states  $|\alpha_n\rangle$  and  $|\beta_n\rangle$ . The case considered includes in fact all known experimental situations. It is clear, that Hamiltonian  $\hat{\mathcal{H}}^{CF}$  of interaction of quantized EMfield with atomic chains can also be represented in the set of variables, which includes the components of spectroscopic transition vector operator  $\hat{\sigma}_v$ . Really, suggesting dipole approximation to be true and polarization of field components to be fixed, we have

$$\hat{\mathcal{H}}^{CF} = -\sum_{j=1}^{n} \sum_{l \neq m} \sum_{m} \sum_{\vec{k}} [p_j^{lm} \hat{\sigma}_j^{lm} (\vec{e}_{\vec{k}} \vec{e}_{\vec{P}_j}) \mathfrak{E}_{\vec{k}} \hat{a}_{\vec{k}} \times e^{-i\omega_{\vec{k}} t + i\vec{k}\vec{r}} + H.c.],$$
(21)

where  $p_j^{lm}$  is matrix element of operator of magnetic (electric) dipole moment  $\vec{P}_j$  of j-th chain unit between the states  $|l_j\rangle$  and  $|m_j\rangle$  with  $l_j=\alpha_j,\beta_j,m_j=\alpha_j,\beta_j,$   $\vec{e}_{\vec{k}}$  is unit polarization vector,  $\vec{e}_{\vec{P}_j}$  is unit vector along  $\vec{P}_j$ -direction,  $\mathfrak{E}_{\vec{k}}$  is the quantity, which has the dimension of magnetic (electric) field strength,  $\vec{k}$  is wave vector,  $\hat{a}_{\vec{k}}$  is field annihilation operator. In the suggestion, that the contribution of spontaneous emission is relatively small, we will have  $p_j^{lm}=p_j^{ml}\equiv p_j$ , where  $l=\alpha,\beta,m=\alpha,\beta$ . Let define the function

$$q_{j\vec{k}} = -\frac{1}{\hbar} p_j (\vec{e}_{\vec{k}} \cdot \vec{e}_{\vec{P}_j}) \mathfrak{E}_{\vec{k}} e^{-i\omega_{\vec{k}}t + i\vec{k}\vec{r}}$$
 (22)

Then the expression (21) can be rewritten in the form

$$\hat{\mathcal{H}}^{CF} = \sum_{v=1}^{n} \sum_{\vec{k}} [q_{j\vec{k}} (\hat{\sigma}_{j}^{-} + \hat{\sigma}_{j}^{+}) \hat{a}_{\vec{k}} + (\hat{\sigma}_{j}^{-} + \hat{\sigma}_{j}^{+}) \hat{a}_{\vec{k}}^{+} q^{*}_{j\vec{k}}], (23)$$

where  $\hat{a}_{\vec{k}}^+$  is EM-field creation operator,  $\hat{a}_{\vec{k}}$  is EM-field annihilation operator, superscript \* in  $q^*_{j\vec{k}}$  means complex conjugation. Field Hamiltonians have usual form

$$\hat{\mathcal{H}}^F = \sum_{\vec{k}} \hbar \omega_{\vec{k}} (\hat{a}_{\vec{k}}^{\dagger} \hat{a}_{\vec{k}} + \frac{1}{2}) \tag{24}$$

for EM-field and

$$\hat{\mathcal{H}}^{Ph} = \sum_{\vec{q}} \hbar \omega_{\vec{q}} (\hat{b}_{\vec{q}}^{\dagger} \hat{b}_{\vec{q}} + \frac{1}{2}) \tag{25}$$

for phonon field. Hamiltonian  $\hat{\mathcal{H}}^{CPh}$  is

$$\hat{\mathcal{H}}^{CPh} = \hat{\mathcal{H}}_z^{CPh} + \hat{\mathcal{H}}_{\pm}^{CPh}, \tag{26}$$

where  $\hat{\mathcal{H}}_{z}^{CPh}$  is determined by the expression

$$\hat{\mathcal{H}}_{z}^{CPh} = \sum_{j=1}^{N} \sum_{\vec{q}} (\lambda_{\vec{q}}^{z} \hat{b}_{\vec{q}} + (\lambda_{\vec{q}}^{z})^{*} \hat{b}_{\vec{q}}^{+}) \hat{\sigma}_{j}^{z}.$$
 (27)

Hamiltonian  $\hat{\mathcal{H}}_{\pm}^{CPh}$  can be represented in the following form

$$\hat{\mathcal{H}}_{\pm}^{CPh} = \sum_{j=1}^{N} \sum_{\vec{q}} \lambda_{\vec{q}}^{\pm} (\hat{\sigma}_{j}^{-} + \hat{\sigma}_{j}^{+}) \hat{b}_{\vec{q}} + (\lambda_{\vec{q}}^{\pm})^{*} (\hat{\sigma}_{j}^{-} + \hat{\sigma}_{j}^{+}) \hat{b}_{\vec{q}}^{+}.$$
(28)

Here  $\lambda_{\vec{q}}^z$  and  $\lambda_{\vec{q}}^\pm$  are electron-phonon coupling constants, which characterisire correspondingly the interaction with z- component  $S_j^z$  and with  $S^+$ - and  $S_j^-$  components of the spin of jth chain unit. It seems to be understandable, that they can be different in general case. Moreover, in order to take into account the interaction with both equilibrium and nonequilibrium phonons both the electron-phonon coupling constants have to be complex numbers, that takes proper account by expressions (27), (28).

It can be shown, that the equations of the motion for spectroscopic transition operators  $\hat{\vec{\sigma}}_l$ , for quantized EM-field operators  $\hat{a}_{\vec{k}}$ ,  $\hat{a}_{\vec{k}}^+$  and for phonon field operators  $\hat{b}_{\vec{q}}$ ,  $\hat{b}_{\vec{d}}^+$  are the following. Instead equation (1) we have

$$\frac{\partial}{\partial t} \begin{bmatrix} \hat{\sigma}_{l}^{-} \\ \hat{\sigma}_{l}^{+} \\ \hat{\sigma}_{l}^{z} \end{bmatrix} = 2 \|g\| \begin{bmatrix} \hat{F}_{l}^{-} \\ \hat{F}_{l}^{+} \\ \hat{F}_{l}^{z} \end{bmatrix} + \|\hat{R}_{\vec{q}l}^{(\lambda^{z})}\| + \|\hat{R}_{\vec{q}l}^{(\lambda^{\pm})}\|, \quad (29)$$

where matrix  $||\hat{R}_{\vec{q}l}^{(\lambda^z)}||$  is

$$||\hat{R}_{\vec{q}l}^{(\lambda^z)}|| = \frac{1}{i\hbar} \begin{bmatrix} 2\hat{B}_{\vec{q}l}^{(\lambda^z)} \hat{\sigma}_l^- \\ -2\hat{B}_{\vec{q}l}^{(\lambda^z)} \hat{\sigma}_l^+ \\ 0 \end{bmatrix}$$
(30)

with  $\hat{B}_{\vec{q}l}^{(\lambda^z)}$ , which is given by

$$\hat{B}_{\vec{q}l}^{(\lambda^z)} = \sum_{\vec{q}} [(\lambda_{\vec{q}l}^z)^* \hat{b}_{\vec{q}}^+ + \lambda_{\vec{q}l}^z \hat{b}_{\vec{q}}]. \tag{31}$$

Matrix  $||\hat{R}_{\vec{a}l}^{(\lambda^{\pm})}||$  is

$$||\hat{R}_{ql}^{(\lambda^{z})}|| = \frac{1}{i\hbar} \begin{bmatrix} -\hat{B}_{ql}^{(\lambda^{\pm})} \hat{\sigma}_{l}^{z} \\ \hat{B}_{ql}^{(\lambda^{\pm})} \hat{\sigma}_{l}^{z} \\ \hat{B}_{ql}^{(\lambda^{\pm})} (\hat{\sigma}_{l}^{+} - \hat{\sigma}_{l}^{-}) \end{bmatrix}, \quad (32)$$

where  $\hat{B}_{\vec{q}l}^{(\lambda^{\pm})}$  is

$$\hat{B}_{\vec{q}l}^{(\lambda^{\pm})} = \sum_{\vec{q}} [(\lambda_{\vec{q}l}^{\pm})^* \hat{b}_{\vec{q}}^+ + \lambda_{\vec{q}l}^{\pm} \hat{b}_{\vec{q}}]. \tag{33}$$

The equation (2) remains without changes. The equation (3) is

$$\frac{\partial}{\partial t} \begin{bmatrix} \hat{b}_{\vec{k}} \\ \hat{b}_{\vec{q}}^{+} \end{bmatrix} = -i\omega_{\vec{q}} ||\sigma_{P}^{z}|| \begin{bmatrix} \hat{b}_{\vec{q}} \\ \hat{b}_{\vec{q}}^{+} \end{bmatrix} + \frac{i}{\hbar} \begin{bmatrix} -\sum_{l=1}^{N} \{\lambda_{\vec{q}l}^{z} \hat{\sigma}_{l}^{z} + \lambda_{\vec{q}l}^{\pm} (\hat{\sigma}_{l}^{+} + \hat{\sigma}_{l}^{-})\} \\ \sum_{l=1}^{N} \{\lambda_{\vec{q}l}^{z} \hat{\sigma}_{l}^{z} + \lambda_{\vec{q}l}^{\pm} (\hat{\sigma}_{l}^{+} + \hat{\sigma}_{l}^{-})\} \end{bmatrix}.$$
(34)

Thus, QED-model for dynamics of spectroscopic transitions in 1D multiqubit exchange coupled system, presented in [14] is generalized by taking into account more carefully quantum nature of relaxation processes. New quantum phenomenon can be predicted. The prediction results from the structure of the equations derived and it means, that by resonance the coherent system of the resonance phonons, that is, the phonons with the energy, egualed to resonance photon energy can be formed, which can lead to appearance along with Rabi oscillations determined by spin (electron)-photon coupling with the frequency  $\Omega^{RF}$  of Rabi oscillations determined by spin (electron)-phonon coupling with the frequency  $\Omega^{RPh}$ . In other words QED-model predicts the oscillation character of quantum relaxation, that is quite different character in comparison with phenomenological Bloch model. Moreover if  $\lambda_{dl}^{\pm} < g$  the second Rabi oscillation process will be observed by stationary state of two subsystems { EM-fied + magnetic (electric) dipoles }, that it will be registered in quadrature with the first Rabi oscillation process. It can be experimentally detected even by stationary spectroscopy methods.

### B. Experiment

ESR studies in anthracite samples have been carried out on stationary ESR spectrometer "Radiopan" at room

temperature. The 100 kHz high frequency modulation of static magnetic field  $H_0$  was used. The static magnetic field  $H_0$  was sweeped in two directions from lesser value to bigger value  $(\frac{dH_0}{dt} > 0)$  and from bigger value to lesser value  $(\frac{dH_0}{dt} < 0)$ . Two regime of registration were used - with automatic microwave frequency adjustment and without automatic microwave frequency adjustment. The spectra obtained are presented in Figures 1, 3-9. The most interesting, that the spectra can be registered by using of 100 kHz high frequency (HF) modulation canal at modulation amplitude  $H_m = 0$ , Figure 1. It is seen from Figure 1, that the spectrum has very unusual for stationary ESR-spectroscopy form, it is similar partly to the spectra, registered by nonstationary transient ESR-spectroscopy. At the same time the functional dependence is another. The spectral distribution of nutation signals in transient ESR-spectroscopy is described usually by Bessel functions. The spectrum, presented in Figure 1, consist of two group of very narrow ESR lines. For instance, the linewidth  $\Delta H$  of the central line with maximal amplitude in left group is equal to  $0.01(\pm 0.004)$  G. It allows to determine q-value (that is g-factor in Zeeman term of spin-Hamiltonian) very precisely. It is equal to  $2.002735 \ (\pm 0.000001)$ . The linewidth of the other lines is practically the same. The lines presented in Figure 1 seem to be the most narrow among all the lines, registered in ESR-spectroscopy at all. The position of the central line with maximal amplitude in right group is given by q-value 2.00255 ( $\pm 0.00001$ ). The structure of the spectrum is very similar to theoretical time evolution for a system with one qubit in JCM, Figure 2. Really, the spectra in both the groups can be described by almost sinusoidal dependence with Gaussian envelope, Figure 3. The qualitative difference between time evolution given by JCM and spectral distribution presented in Figure 1 is that, the left group of lines in JCM has cosinusoidal dependence, Figure 2. The possibility to explain the spectrum observed by usual amplitude modulation is excluded, since the envelope by amplitude modulation is described by sinusoidal dependence, we have the envelope, described by Gauss distribution with slightly different parameters for left and right sides. So the fit of experimental data, Figure 3, for left hand side group of lines gives the following expression

$$y = y_0 + \frac{A}{W} \sqrt{\frac{2}{\pi}} \exp\{-2\left[\frac{(H - H_c)}{W}\right]^2\},$$
 (35)

where  $y_0 = 52.4$ ,  $H_c = 3322.95G$ , W = 0.36G, A = 32G.

Given result and comparison with Figure 2 allow to conclude that the spectrum in Figure 1 represents itself the quantum Rabi oscillation picture. Really, let us touch on JCM in some details. Central role in JCM plays the sum with infinite summation limit, which represents on a scale [-1,1] the degree of excitation of two-level system resulting of interaction between the atom dipole or spin

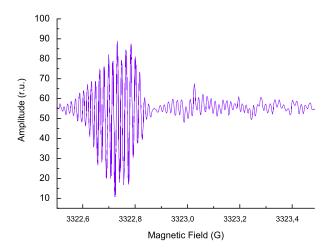


Figure 1: ESR spectrum of anthracite sample, registered by 100 kHz high frequency modulation of static magnetic field with amplitude  $H_m=0$ 

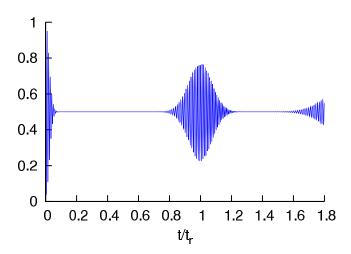


Figure 2: Theoretical time evolution for a system with one qubit in JCM, initial state  $|\psi\rangle\otimes|\alpha\rangle$  is direct product of atomic ground state and coherent field state with  $\overline{n}=50$ 

and single mode quantized EM-field. It is

$$\langle \hat{\sigma}^z(t) \rangle = -\exp[-|\alpha|^2] \sum_{n=1}^{\infty} \frac{|\alpha|^{2n}}{n!} \cos 2g\sqrt{\overline{n}}t,$$
 (36)

where  $|\alpha\rangle$  is fully coherent state of the field, taking place at t=0, at that  $|\alpha|^2=\overline{n}$ , that is, it is the average number of photons in the field, g is coupling constant between field and atom (spin). The sum in (36) cannot be expressed exactly in analytical form. For very short times and very large  $\overline{n}$  behavior of  $\langle \hat{\sigma}^z(t) \rangle$  is determined by  $\cos 2g\sqrt{\overline{n}}t$ . Cummings [24] has shown, that by resonance and for intermediate time t values the cosine Rabi oscillation damp quickly (so called collapse takes place).

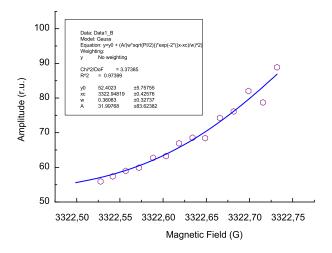


Figure 3: The fit of left hand side envelope of left oscillation group in the spectrum, presented in Figure 1 by Gaussian

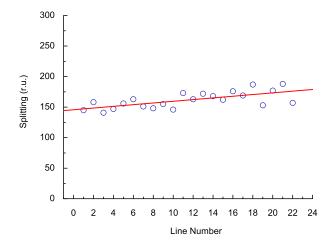


Figure 4: The fit of the splitting between the lines in left oscillation group in the spectrum, presented in Figure 1 by linear dependence

Given damping can be described by Gaussian envelope

$$\exp[-\frac{1}{2}(gt)^2].$$
 (37)

It is substantial, that it not depends on field intensity unlike to semiclassical Rabi oscillation damping process and it is determined entirely the only by coupling constant g. The authors of the work [25] have found, that JCM contains so called revival process with revival time  $T_R$ , given by the expression

$$T_R = \frac{\pi}{a^2} \sqrt{\Delta^2 + 4g^2 \overline{n}},\tag{38}$$

where  $\Delta$  is deviation of field mode frequency from res-

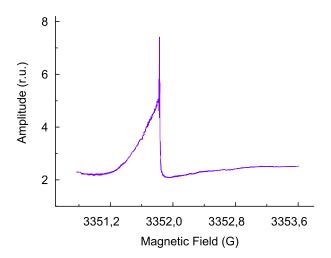


Figure 5: ESR spectrum of anthracite sample, registered by 100 kHz high frequency modulation of static magnetic field with amplitude  $H_m=0.01G$  and with automatic microwave frequency adjustment,  $\frac{dH_0}{dt}<0$ 

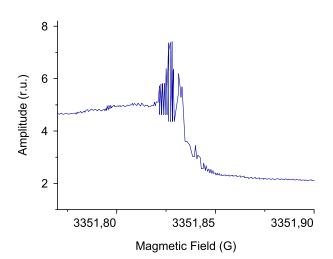


Figure 6: Central part of ESR spectrun of anthracite sample, registered by 100 kHz high frequency modulation of static magnetic field with amplitude  $H_m=0.01G$  and with automatic microwave frequency adjustment,  $\frac{dH_0}{dt}<0$ 

onance value. Revival process takes place at all time values, satisfying the relation  $t=kT_R, k\in N$ . It is seen from (38) that revival time depends on  $\overline{n}$  and it is proportional to field amplitude at  $\Delta=0$  like to dependence of Rabi frequency by semiclassical consideration. Let us illustrate the dynamics of spectroscopic transitions in JCM. The initial state for single qubit system can be defined by direct product of the two level matter (atomic, spin and so on) subsystem state  $|\psi\rangle$  and coher-

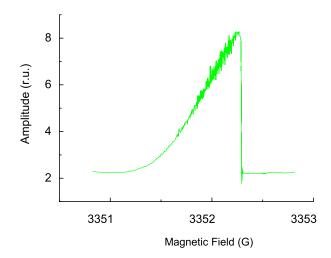


Figure 7: ESR spectrum of anthracite sample, registered by 100 kHz high frequency modulation of static magnetic field with amplitude  $H_m=0.01G$  and with automatic microwave frequency adjustment,  $\frac{dH_0}{dt}>0$ 

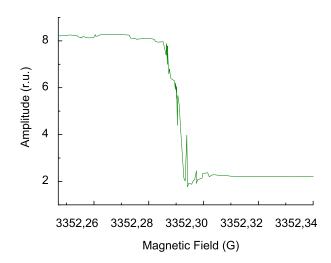


Figure 8: Central part of ESR spectrun of anthracite sample, registered by 100 kHz high frequency modulation of static magnetic field with amplitude  $H_m = 0.01G$  and with automatic microwave frequency adjustment,  $\frac{dH_0}{dt} > 0$ 

ent EM-field state  $|\alpha\rangle$ . It is the following

$$|\Psi(0)\rangle = |\psi\rangle \otimes |\alpha\rangle,\tag{39}$$

where  $|\psi\rangle$  is

$$|\psi\rangle = c_1|\psi_1\rangle + c_2|\psi_2\rangle \tag{40}$$

and  $|\alpha\rangle$  is

$$|\alpha\rangle = \exp\left[\frac{-|\alpha|^2}{2}\right] \sum_{n=1}^{\infty} \frac{|\alpha|^n}{n!} |n\rangle, \alpha = \sqrt{\overline{n}} e^{i\phi}.$$
 (41)

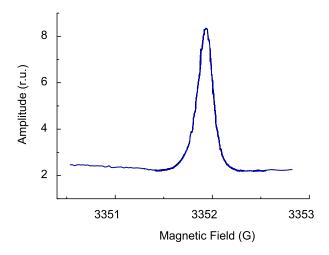


Figure 9: ESR spectrum of anthracite sample, registered by 100 kHz high frequency modulation of static magnetic field with amplitude  $H_m=0.01G$  and without automatic microwave frequency adjustment,  $\frac{dH_0}{dt}>0$ 

The Rabi oscillations of the probability, that the qubit is in the initial state in the first damping stage (collapse), on a time scale of  $t_c \simeq \frac{\sqrt{2}}{g}$  and then revive at  $T_R \simeq \frac{2\pi\sqrt{\overline{n}}}{g}$  are illustrated in Figure 2 for  $c_1 = 1$ ,  $c_2 = 0$  by plotting

$$\sum_{n=1}^{\infty} |\langle \psi_1, n | \Psi(t) \rangle|^2, \tag{42}$$

where  $\langle \psi_1, n | = \langle \psi_1 | \otimes \langle n |$  is the ground state for the qubit with n photons in the cavity. Our measurements of the dependence of the ESR spectral characteristics on the microwave power (photon number) is also showed that in correspondence with JCM the positions of the lines of left hand side group are not dependent on photon number, at the same time the positions of the lines of right hand side group are dependent on photon number. The aforesaid difference between our results and JCM can be easily explained if we determine the Rabi frequency value. It is equaled to 28 kHz. Given value is substantially lesser in the same microwave power range, than the Rabi frequency values in excited by microwave power systems. So, for instance, in Si the Rabi frequency changes from  $\approx 1 \times 10^6 rads^{-1}$  to  $\approx 4.5 \times 10^6 rads^{-1}$ for the Si-P3 centers [26]. It means that we really have observed the quantum relaxation oscillations, predicted theoretically in Section 2A. The prediction of sinusoidal character (instead cosinusoidal in JCM) is also confirmed. The comparison with the model for dynamic of spectroscopic transitions, given in Section 2A is correct, since at modulation amplitude  $H_m = 0$  the absorption cannot be registered. At the same time the emission can be registered, since by detection at 100 kHz the quantity which changes essentially slowly is registered like to usual ESR detection. Then, if to omit the part from Hamiltonian,

corresponding to EM-field and z-relaxation term and to set N equaled to 1, we will have in rotating wave approximation JCM Hamiltonian for phonon subsystem. The linear dependence of splitting, presented in Figure 4, can be explained by the dependence of  $\Omega^{RPh}$  on the static H-field, since by the change of static H-field the distance between the energy levels in qubits is also changes. Expanding the dependence  $\Omega^{RPh}(H)$  in Taylor series and restricting by linear term (it is correct, since H-change is small), we obtain the agreement with experiment.

To confirm the conclusion on coherent emission character (which is like to maser emission character) of the spectrum, observed by  $H_m = 0$  and to establish the structure of the centers, which can emit stationary, we have undertaken the study of ESR-responce at various values of HF-modulation amplitude. The spectra, registered by 100 kHz HF-modulation of static magnetic field with amplitude  $H_m = 0.01G$  are presented in Figures 5 to 9. The lines have also unusual shape for absorption derivative. The spectra are also very different by the change of sweep direction, compare Figures 5 and 7, 6 and 8, which all were registered with automatic microwave frequency adjustment. Given difference disappears by registration without automatic microwave frequency adjustment and for both sweep directions we obtain the spectrum, presented in Figure 9. It is substantial, that observed polarity of derivative of ESR responce by usual registration with automatic microwave frequency adjustment does not depend on sweep direction. At the same time, when ESR spectra by  $\frac{dH_0}{dt} > 0$  and by  $\frac{dH_0}{dt} < 0$  are absorbtion spectra the polarities of qiven spectra have to be different [27]. Consequently one of the spectrum is emission spectrum. It is evident, taking into consideration the energy conservation law, that emission is observed by  $\frac{dH_0}{dt} < 0$ . Although the emission is coherent, it is not maser effect, since the amplification is absent (it seems to be in principle impossible for two level system). Then the hysteresis effect (compare Figure 5 and 7), which is observed by registration with automatic microwave frequency adjustment can be explained in a natural way like to Stoks luminescence, where the emission spectrum is shifted to more low frequences in comparison with excitation spectrum to be consequence of interaction with nonresonance phonons. The key to structure of ESR centers can be found by the analysis of the central part of ESR spectra by registration with automatic microwave frequency adjustment, presented in Figures 6 and 8. It is seen from Figure 6, that Rabi oscillation process is also takes place, although spectral resolution is not very good, since the modulation amplitude is coinciding with linewidth. The most interesting is the shape of background line. In both the spectra the shape of background line has kink form and can be be approximated by tanh-function. It can mean, that the model of the ESR centers has to be similar to Su-Schrieffer-Heeger (SSH) model of topological solitons in trans-polyacetylene [28]. The g-factor value, which is very near to g-factor value of neutral solitons in trans-polyacetylene testifies in favour of given proposal.

It can mean, that structural element of anthracite includes carbon backbone of trans-polyacetylene. At the same time anthracite is structurally disordered material. It means, that the solitons has to be pinned and they have to give usual ESR spectra. To explain the appearance of kinks, at that it is substantionally, that they are characterized by different parameters for absorption and emission spectra, and the possibility to register Rabi oscillations, we have to consider joint {EM-field + magnetic dipole + resonance phonon field} system. EM-field and resonance phonon field produce radiation communication in the direction of EM-field propagation and in time. It means that we have 1D chain, at that the chains of two kinds are produced in time - with the sequence - absorption time segment - emission time segment and with reversal sequence. It is substantionally, that given time segments are different in their values. In fact the dimerisation take place like to trans-polyacetylene, however instead space z-axis along t-axis. It is in agreement with known equality of rights of coordinates in Minkowski space. In other words the time has to be considered being to be quantized in given case. The junction of given time ordered chains produces time topological violations of two kinds. The first topological violation can effectively absorb photons and emit resonance phonons, the second topological violation has reversal properties. So, we come to the model of instanton, which is like to some extent to SSH-soliton model. It is understandable, that the instantons of both kinds can free, that is without additional activation energy, move along t-axis like to SSHsolitons along space z-axis. It is the first experimental detection of instantons (to our knowledge). The known instantons in the literature are considered to be a special kind of vacuum oscillations in gluon field theory and also in gravitation field theory [29]. They have quite another origin.

Therefore we have observed new kind of coherent states. They are modes of resonance phonon field. The coherence time value in the samples studied is evaluted to be equal at room temperature  $\sim 10ms,$  at that at relativelly low microwave power and it will increase with power increase, achieving the maximal known values at present and even more. Substantial advantage for possible applications is that, that the responce on external EM-field is very strong. At the same time the detection of single spin is rather technically difficult task. The H-field dependence allows to separate the phonon modes, the dependence on H-field direction seems to be also essential for practical applications.

### III. CONCLUSIONS

QED-model for dynamics of spectroscopic transitions in 1D multiqubit exchange coupled system, presented in [14] is generalized by taking into account more carefully quantum nature of relaxation processes. It is predicted, that by resonance the coherent system of the resonance phonons, that is, the phonons with the energy, equaled to resonance photon energy can be formed. It leads to appearance along with Rabi oscillations determined by spin (electron)-photon coupling with the frequency  $\Omega^{RF}$  of Rabi oscillations determined by spin (electron)phonon coupling with the frequency  $\Omega^{RPh}$ . Therefore QED-model predicts the oscillation character of quantum relaxation, that is quite different character in comparison with phenomenological Bloch model. Moreover if  $\lambda_{\vec{a}l}^{\pm} < g$  the second Rabi oscillation process will be observed by stationary state of two subsystems {EM-fied + magnetic (electric) dipoles, that means, that it will be registered in quadrature with the first Rabi oscillation process. It can be experimentally detected even by stationary spectroscopy methods. All the predictions are experimentally confirmed. The model of new kind instantons is proposed.

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